Development of Benzo[*de*]isoquinolino[1,8-*gh*]quinoline Diimide n-Type Organic Semiconductors with Asymmetric Substituents

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The molecular assembly of organic semiconductors (OSCs) is arguably the most essential factor to achieve band-like charge-transport[1], suppression of molecular flucuations[2], and high charge-carrier mobility (μ). In contrast to the high-performance hole-transporting p-type OSCs (μ > 10 cm^2 V^{-1} s^{-1})[3], the electron-transporting n-type OSCs exhibit much lower μ that are inadequate for the construction of all-organic logic circuits. It shows that our current understanding of molecular assemblies needs to be improved in order to effectively develop high-performance n-type OSCs. Recently, our group developed a robust synthetic method to decorate the nitrogen-containing benzo[*de*]isoquinolino[1,8-*gh*]quinoline diimide (**BQQDI**)[4] π-electron core with asymmetric substituents (**Figure**). We have demonstrated that the interlayer interactions of asymmetric **BQQDI** can be effectively tuned by varying the length of alkyl sidechains, and we also discovered an intriguing *gauche* alkyl conformation as a result of molecular mimicry to achieve favorable molecular assemblies and suppressed molecular fluctuations. To further examine the molecular assembly-performance relation of **BQQDI**, we investigated the effect of different heterocyclic substituents in asymmetric **BQQDI**. Owing to the labile nature of some heterocycles, a new synthetic strategy with mild reaction conditions was developed, and the asymmetric **BQQDI** derivatives were extensively investigated in terms of molecular assemblies, charge-transport properties, and device performances to advance our understanding of high-performance n-type OSCs.

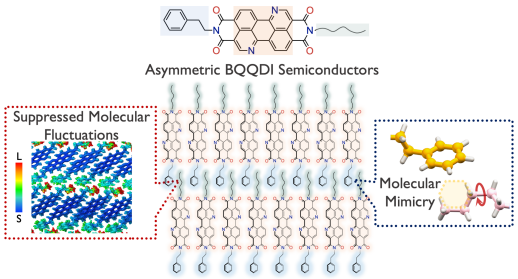

Figure. Molecular design of asymmetric **BQQDI**.

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